TRANSPARENT CONDUCTING OXIDE ELECTRODES REQUIREMENTS FOR HIGH EFFICIENCY MICROMORPH SOLAR CELLS

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ABSTRACT: The requirements for a micromorph tandem cell front transparent conductive oxide (TCO) are multiple. This essential layer needs a high transparency, excellent conduction, strong light scattering into silicon and good surface morphology for the subsequent growth of silicon cells. These parameters are all linked and trade-offs have to be found for optimal layer. The optimum combination, taking into account current achievable materials properties, is still unclear. Concerning transparency, we study here the impact of free carrier absorption (FCA) on the photogenerated current by using first doped and non-intentionally-doped zinc oxide (ZnO). Then, Bi-layers made of flat indium tin oxide (ITO) under various thicknesses of rough ZnO allow a study of the haze influence alone. It is shown that FCA induces drastic current losses in the infra-red part of the spectrum, and haze increase enhances the cell response in the infra-red part up to a certain limit of grain size. Surface feature sizes above 0.4 μ m appear to be useless for haze increase purpose at the ZnO/Si interface. By using an optimized 2 μ m thick LPCVD ZnO, micromorph cells showing 13.7% initial efficiency, with a total current of 27.7 mA/cm² could be obtained with 240nm and 2.8 μ m of top and bottom cell thicknesses.

Keywords: Thin Film Solar Cell, Light Trapping, TCO Transparent Conducting Oxides

1 INTRODUCTION

1.1 General context

The micromorph tandem cell concept, comprising a hydrogenated amorphous silicon (a-Si:H) top cell and a hydrogenated microcrystalline silicon (μ c-Si:H) bottom cell, is promising for low-cost photovoltaic energy [1]. The main challenge to achieve high efficiency with such devices is to develop a cell design allowing for high currents in both cells while maintaining optimum electrical properties.

One key element in that respect is the front electrode which needs to exhibit high transparency and conduction, strong light scattering at the textured interface to silicon for optimum light trapping in the cell [2-4]. Conduction and transparency are mainly controlled by the density and mobility of free carriers, as they assure the conduction and determine the near-infrared absorption (free carriers absorption, FCA). Light scattering potential is mainly defined by the haze in transmission (ratio of diffusively transmitted light over total transmitted light) and its angular distribution. Understanding the respective influence of FCA and of light scattering on the current of the cell is not trivial [4, 5]. This is what we intend to realise in this work, in order to design the optimal TCO. We show that on a $2\mu m$ thick low pressure chemical vapour deposited (LPCVD) ZnO total currents over 28mA/cm² can be reached.

1.2 Experimental details

The devices presented in this study are micromorph tandem cells deposited on textured ZnO deposited on glass by low pressure chemical vapor deposition (LP-CVD). The ZnO is then slightly treated with a surface Ar plasma to round the shape of the pyramids. When used (in section 3), flat ITO is deposited by sputtering on glass before ZnO. Silicon layers are deposited by plasma enhanced chemical vapor deposition (PECVD). The micromorph cells include a silicon oxide based intermediate reflector (SOIR) [6].

The cells are patterned 1 cm^2 by lift-off and dryetching. LPCVD ZnO and a white dielectric are used as back electrode and back reflector. The current-voltage (*I*-*V*) curves of the cells are measured with a dual lamp WACOM solar simulator in standard test conditions (25°C, AM1.5G spectrum, 1000 W.m⁻²) and open-circuit voltage (V_{oc}) and fill-factor (*FF*) are calculated. The short-circuit current density (J_{sc}) is determined by integrating the product of the external quantum efficiency (EQE) and the incoming photon flux of the AM1.5G spectrum.

2 FCA EFFECT ON THE CELL CURRENT

2.1 Different electrodes

Fig. 1a presents schematic side views of three micromorph cells with identical silicon layers but different front and back TCO electrodes. In all three cases, front and back TCO are similar. In case A, an industrial-like 2μ m thick boron-doped LPCVD ZnO single layer with a rough surface (70nm rms roughness) and a sheet resistance lower than 10 Ω /sq is used for reference.

The same morphology is reproduced in case B with non-intentionally-doped (n-i-d) ZnO but with a 1.6µm thick film. A thinner layer is needed when no extrinsic dopant is added because of changing growth condition [2]. It presents the advantage of a much lower FCA thanks to a carrier density around 2.10¹⁹ cm⁻³ (7 times lower than for the previous case) [7]. Its absorptance, as well as that of the industrial-like aforementioned TCO, can be seen in Fig. 1b. Such a film shows however a much higher sheet resistance close to $40\Omega/sq$. To keep the advantage of a low free carriers density without changing the surface morphology but with a $10\Omega/sq$ sheet resistance, one needs to use a thick, flat n-i-d LPCVD ZnO under the 1.6µm thick n-i-d ZnO film (e.g. a 7µm thick film as in the scheme C). Such a layer can be obtained by chemical mechanical polishing. A very thin interlayer (like a 3nm thick n-type microcrystalline silicon layer) is used in this case to avoid an epitaxial growth of the second ZnO layer.

2.2 Resulting absorption and currents

Fig. 1b shows the absorptance of the TCO front electrodes of A, B and C devices. The lower carriers density of n-i-d ZnO makes the absorptance in the B case much lower than A. The much larger thickness in the C case induces stronger absorptance than in A and B cases for the short wavelengths range. However, above 550nm, the absorptance is lower than that of the A case (thanks to higher transmittance in the near infra-red due to lower doping [7]).



Figure 1: a) Schematic view of 3 micromorph cells with different front and back TCO based electrodes b) Absorptance of all 3 front electrodes. c) External quantum efficiency of top and bottom cells of the three devices presented in a) (plain lines). Total EQE (dotted lines) as well as "1-reflectance" (dashed lines) are indicated as well. The legend indicates for all 3 devices the top and bottom cell current in mA/cm².

Fig. 1c presents the EQE of top and bottom sub-cells of micromorph devices in cases A, B and C. The lower absorptance of thin electrodes (A and B) in the short wavelengths enhances the top cell response compared to case C. However, the low photon density in the solar spectrum for these wavelengths keeps the difference in top cells currents quite low (around 0.1mA/cm²). The EQE curves of top and bottom cell cross very close to each other confirming an identical light scattering capability for all 3 front electrodes.

The bottom cell response for case A is drastically lower than other cases because of the increased FCA due to higher free carriers density. 2mA/cm^2 can be gained when using n-i-d ZnO instead of doped ZnO for the same order of thicknesses (at the cost of an increased sheet resistance). Moreover, for the same $10\Omega/\text{sq}$ sheet resistance, using n-i-d ZnO front and back electrodes (around $8\mu m$ thick) instead of doped ones (yet 4 times thinner) permits to gain 1.4mA/cm^2 . This proves the paramount importance of reducing as much as possible the carriers density in LPCVD ZnO films used as TCO electrodes in devices including microcrystalline silicon.

3 HAZE INFLUENCE ON THE CELL CURRENT

3.1 Different front electrodes

To study the impact of changing the front electrode haze on micromorph cells current, micromorph cells are now deposited on ITO-ZnO bi-layer front electrodes. The first 60nm thick ITO layer ensures good conduction for all cases; moreover, its absorptance in the near infrared (due to high free carriers density around 7.10^{20} cm⁻³) is more than 30 times that of n-i-d ZnO and induces thus an very similar total absorptance for all electrodes. The ZnO thickness is changed from 0.1µm to 7µm to increase the pyramidal feature size on its surface (e.g. for 1µm and 7 µm thick films in Fig. 2a). This results in an enhancement of the light scattering (in particular the haze) as shown in Fig. 2c [2]. We kept the angular dependence of the light scattering identical for all thicknesses, as can be seen with angular distribution functions of scattered light curves in the Fig. 2b.



Figure 2: a) SEM images of LPCVD ZnO surface for 1μ m and 7 μ m thick layers. b) normalized angular distribution function of scattered light for 0.1μ m to 7μ m thick ZnO films. c) spectral dependences of haze in transmission of these layers measured in air. The spectral dependence of the haze calculated in silicon for 1μ m and 2μ m thick films is plotted in dashed lines.

3.2 Resulting effects on the cells currents

Fig. 3 presents the EQE of top and bottom sub-cells of micromorph devices grown on aforementioned front electrodes. It can be seen in the legend that increasing roughness (i.e. ZnO thickness in our case) of front electrodes enhances the bottom cell current but that the top cell current shows a maximum for a 2μ m thick film. The continuous gain in bottom cell current can be attributed to better light scattering, reduced absorption in doped layers and weakening of the reflection from the SOIR [8]. This last point explains also the reduction of top cell current for ZnO films thicker than 2μ m.

All these points result in enhancement of the bottom cell EQE when increasing the front ZnO thickness (i.e. the haze). However, changes in the intermediate reflection due to the SOIR are visible in the 500nm – 850nm range, while light scattering changes have a bigger impact for larger wavelength. EQE at 900nm wavelength and higher values is thus more relevant to evaluate the light scattering impact.

Noteworthy, EQE curves of cells grown on 2µm,

 $4\mu m$ and $7\mu m$ thick films are superimposed after 900nm. This indicates that $2\mu m$ thick ZnO films appear to show sufficient light scattering ability. Actually, a simple calculation of the haze from this $2\mu m$ thick film into silicon [9] is close to 1 for all wavelengths up to 1100nm, which is not the case for a $1\mu m$ thick ZnO film (see dashed lines in Fig. 2c). For a given wavelength, when increasing surface feature size (as defined in [7] as dimensional parameter or grain size) value, the haze of TCO based rough layers into silicon reaches unity before the one measured in air.



Figure 3: External quantum efficiencies of top and bottom subcells of micromorph devices grown on ITO-ZnO bi-layers with various ZnO thicknesses. Top and bottom sub-cells currents in mA/cm² are indicated in the legend.

Moreover, the value in silicon is much more relevant to characterize light scattering in the cell as can be observed in Fig. 4: when increasing the surface feature size (i.e. the thickness of the layer for LPCVD ZnO), EQE taken at the same wavelength as the haze also saturates to a maximal value; this saturation occurs for surface feature sizes in the same order as the saturation of haze into silicon. Yet, for ZnO layers with such structures, haze measured in air remains around 50%.



Figure 4: Right axis: Haze in transmission measured in air and calculated in silicon for 900nm wavelength incident light for various thicknesses of LPCVD ZnO front electrodes as a function of the typical size of their surface features. Left axis: EQE value at the same wavelength of micromorph cells grown on the same LPCVD front electrodes.

Thus, 2μ m thick ZnO, which gives on its surface pyramidal shapes of 0.4 μ m typical size, already gives maximal light scattering capabilities for microcrystalline silicon and also gives the best top cell response. It thus appears to be quite optimal for high matched currents in micromorph devices.

4 OPTIMIZED DEVICES

Micromorph cells having a higher V_{oc} and lower J_{sc} than amorphous cells, they can use TCO electrodes with quite high sheet resistances with reduced efficiency losses [10]. It is thus possible to use TCO with sheet resistances higher than 10 Ω /sq as front and back electrodes. When optimized, a simple 2µm thick n-i-d ZnO layer can have more than 50cm².V⁻¹.s⁻¹ of mobility with still around 2.10⁻¹⁹ cm⁻³ of carrier density, resulting in sheet resistances around $30\Omega/sq$ after the cell deposition. The small FF loss due to this increased sheet resistance compared to the classically admitted $10\Omega/sq$ is overcompensated by the current gain from the reduced absorptance of this layer. A 1cm² micromorph cell using doped nano-crystalline silicon oxide layers [11, 12] showing 13.7% initial efficiency could be obtained on such a front electrode with 1.38V, 13.8mA/cm² (total current of 27.7 mA/cm²) and 72% of V_{oc} , J_{sc} and FF respectively. IV curve and EQE of this device (240nm top and 2.8µm bottom cell thicknesses) are shown in Fig. 5.



Figure 5: EQE curve inside the IV curve of an optimized 1 cm2 micromorph cell using $2\mu m$ thick n-i-d ZnO front electrode deposited on AR coated glass. Top / bottom cell thicknesses are $240 nm / 2.8\mu m$.

5 CONCLUSION

In summary, a large bottom cell EQE increase, thus a current gain are observed when using TCO with low free carriers density thanks to reduced FCA. On the other hand, enhancing the haze also improves the bottom cell current, yet high hazes measured in air are not mandatory for good light scattering in silicon, and 2μ m thick LPCVD ZnO films with a typical feature size of 0.4 μ m are sufficient to reach the maximal gain in EQE at large wavelength. Finally, high efficiency micromorph cells could be done on 2μ m thick n-i-d LPCVD ZnO. It shows excellent light scattering properties (for both top and bottom sub-cells) and low absorptance that are advantages to reach high top and bottom cell currents.

6 ACKNOWLEGEMENT

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